Micro-Raman analysis of molar fraction in polycrystalline $In_xGa_{1-x}As$ for traveling liquidus zone growth method

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$In_xGa_{1-x}As$ bulk crystals are promising lattice-matched substrate materials for InGaAs-based optoelectronic devices. By choosing a suitable value of the molar fraction x, $In_xGa_{1-x}As$ substrates can provide superior lattice-matching between the epilayer and the substrate, compared to the conventional InP substrates. A wide variety of devices can be designed on a multi-component InGaAs single crystal, for which an important requirement becomes the homogeneity of the molar fraction across the whole sample. Attempts to grow homogeneous $In_xGa_{1-x}As$ material, particularly with high value of x, have remained unsuccessful. Under micro-gravity conditions [1], it is expected that the growth of single crystal with homogeneous composition would be easier, as the convective flow is suppressed in such conditions. However, the growth qualities are still limited by the weak existing melt convection and residual acceleration. Our initial studies indicate that this drawback can be overcome by using polycrystalline InGaAs starting material with a certain graded molar fraction, which compensates the effect of weak melt convection.

It is planned to grow compositionally homogeneous $In_xGa_{1-x}As$ bulk substrate under micro-gravity conditions using $In_xGa_{1-x}As$ polycrystals with graded molar fraction as the starting material in the traveling liquidus zone method [2]. The growth will be carried out using the vertical Bridgeman (VB) technique, where the starting material should have a cylindrical shape. A particular gradient of molar fraction in this starting material is a very critical requirement, and therefore it is important to analyze the molar fraction in this starting material using a powerful accurate method. In this paper, we present our studies on the starting material. As the first step, we have grown $In_xGa_{1-x}As$ polycrystals with various molar fraction. In the next step, we have grown polycrystalline $In_xGa_{1-x}As$ with graded molar fraction, which was grinded into cylindrical shape. In a view to analyze the molar fraction in these polycrystalline starting materials using a non-destructive method, we report some results of micro-Raman scattering studies on these samples. Results obtained from Raman scattering are compared with those obtained from the conventional method of chemical analysis, which is destructive in nature. An additional advantage of micro-Raman technique is that it can be used to estimate the molar fraction at a spatial resolution of a few microns. For the cylindrical sample, the results are compared for as-grinded and polished surface conditions.

The optical phonons in Raman spectra of compound semiconductors show a composition-dependent shift, which was first observed in infrared reflection experiments [3], provides a convenient non-destructive way to estimate the molar fraction with high accuracy. The theoretical dependence between molar faction and phonon position for $In_xGa_{1-x}As$ can be given by Eq. (1). Figure 1 presents the first-order optical phonons in Raman spectra of some of the polycrystalline InGaAs samples, where a composition-dependent shift can be observed. Various phonons were deconvoluted using lineshape fitting, as shown in the inset. Figure 2 shows the experimental values of the phonon frequency and the theoretical lines obtained from Eq. (1). By comparing the experimental values with theoretical lines, the molar fraction is estimated. Molar fraction was also obtained independently from chemical analysis method. The inset of Fig. 2 shows good agreement between the two results. In the case of cylindrical sample, Raman spectra had poor quality for the as-grinded surface compared to the polished surface. Some additional structures were seen in Raman spectra from as-grinded surface, probably due to oxide formation on the surface during the grinding process, which were removed after polishing. However, the estimated values of molar factions were closely identical. Figure 3 shows the molar faction gradient along the length of the cylindrical sample estimated from micro-Raman scattering before and after surface polishing. Our study confirms that micro-Raman technique is one of the best way for accurate routine characterization of molar fraction with high spatial resolution compared to other low-accuracy or destructive methods.

[1] S. Fujiwara, M. Tatsumi, T. Araki, M. Irikura, K. Matsumoto, S. Murai, and T. Shirakawa, J. Associ. Crystal Growth **21** (1994) 411.
[2] K. Kinoshita, H. Kato, M. Iwai, T. Tsuru, Y. Muramatsu and S. Yoda, J. Crystal Growth (in Press, 2000)
[3] M. H. Brodsky and G. Lucovsky, Phys. Rev. Lett. **21** (1968) 990.

Equations

$$
\begin{aligned}
\omega(\mathrm{LO_{GaAs}}) &= 291 - 53 \cdot x, \qquad (1)\\
\omega(\mathrm{TO_{GaAs}}) &= 268 - 30 \cdot x,\\
\omega(\mathrm{LO_{InAs}}) &= 230 + 8 \cdot x,\\
\omega(\mathrm{TO_{InAs}}) &= 230 - 10 \cdot x.
\end{aligned}
$$

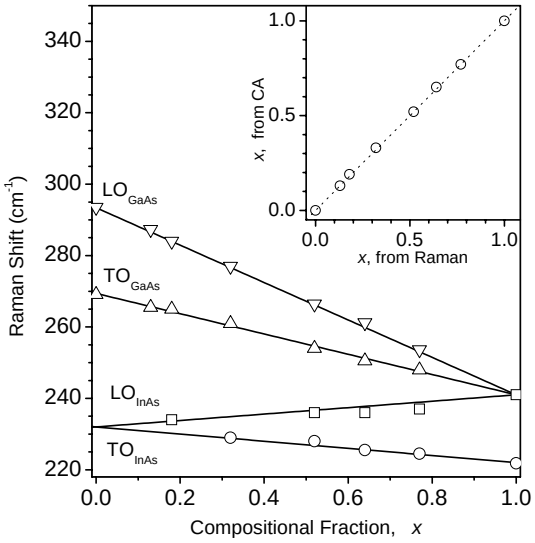

Figure 2: Experimental points and calculated curves for the shift in various phonon frequencies with respect to the composition x. The inset shows a comparison of the results obtained from Raman scattering with those obtained from chemical analysis.

Figures

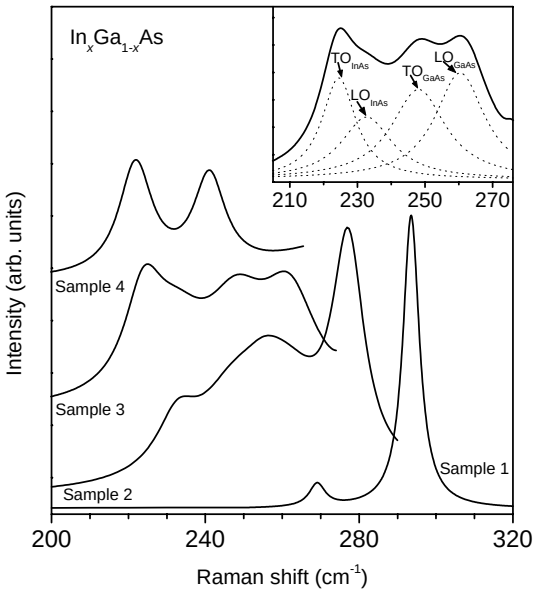

Figure 1: Raman spectra obtained from some of the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ polycrystalline samples. The inset shows line shape fitting for the spectrum from the sample S6.

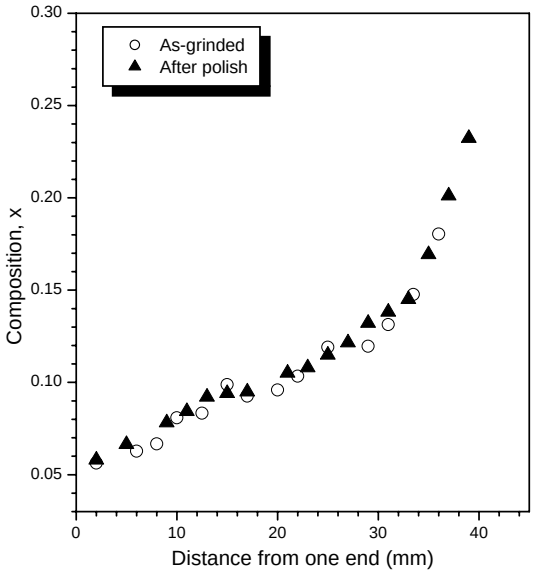

Figure 3: Estimated values of the composition x along the length for the cylindrical sample. The open circles indicate results from the as-grinded surface condition and the full triangles correspond to the results after surface polishing.